“Interactive comment on “Laser induced fluorescence based detection of atmospheric nitrogen dioxide and comparison of different techniques during the PARADE 2011 field campaign” by Umar Javed et al.”

Response: Umar Javed et al.

“Anonymous Referee #1”

“General comments:”

“This manuscript describes a new laser induced fluorescence instrument developed for ground-based and aircraft measurements of NO2. The authors report the instruments characteristics, laboratory tests, an extensive description of the calibration system, and the first results of a field campaign in 2011, where they carried out an intercomparison with other systems that measured NO2 using different techniques. The manuscript is generally well written, and the main results of the intercomparison and the description of a new LIF system characteristics, which uses a CW laser is of a certain importance for future developer of NO2 systems and for the all community working on NOx measurements. In my opinion it fits with AMT scopes and I recommend publication, after the authors address the following questions and comments.”

Response: We are thankful for the Anonymous Referee (#1) for the review and useful comments on the draft.

“Specific comments:”

“**Lines 52-67:** Since the aim of the manuscript is to describe a new NO2 instruments, and because there are different techniques to measure NO and NO2, I would limit the review of the measurements techniques to those for NO2 observation, omitting those for NO detection.”

Response:

We agree with the reviewer. Lines 55-67 (based on the discussion draft) are replace in the revised draft as follows

“The Photofragmentation Two-Photon Laser-Induced Fluorescence (PF-TP-LIF) (Sandholm et al., 1990;Bradshaw et al., 1999) and chemiluminescence (Fontijn et al., 1970) methods are well known for direct in situ NO detection. In the past, an indirect detection of NO_2 with these techniques has been performed by converting $NO_2 \rightarrow NO$ via photolytic/catalytic process followed by NO detection. However, in the case of NO_2 to NO conversion, a potential interference from NO_z species cannot be fully excluded for the NO_2 measurement, e.g. (Crawford et al., 1996;Villena et al., 2012;Reed et al., 2016).”

“**Line 150-155:** I suggest to describe with more details the time-resolved fluorescence signal detection, trigger system, synchronization, how to take care of laser power fluctuation and so on, since this is the key part of the system that may be managed carefully using a CW laser.”

Response:

To explain the data acquisition following “sentences” are included in the draft.

“A counter card is used for the data acquisition. There is no need for synchronisation as the counter card itself triggers the laser pulse. The timing system is entirely controlled by an FPGA (field-programmable gate array), utilizing an external crystal oscillator of 20MHz nominal frequency with a stability of +/-2.5ppm over the temperature range of -30°C to +75°C. All internal frequencies are derived from this clock by means of a PLL (phase-locked loop) in the FPGA. The triggering occurs at a fixed rate of 5 Mhz. The delay caused by the length of the trigger cable (propagation delay of the pulse), the laser power supply unit, propagation delays from detector to FPGA, etc. is compensated with a programmable delay for the data acquisition in the FPGA.

So the FPGA logic recognizes when it should start recording the data after it emitted the trigger pulse and waits the specified amount of programmed clock cycles after emitting the trigger.”

The signal from the PMT is attained for both periods of the laser cycle (100 ns ON, 100 ns OFF). The card has more than 50 channels available for the PMT data. Each channel has a resolution of 4 ns. The integrated raw data is written in a bin file for the sampling period. The sampling period is typically 1s. The NO_2 fluorescence signal is resolved in the post analysis of the raw data. To elaborate the raw signal, a figure for the time-resolved fluorescence is added as a subpart of ‘Fig. 2’to the draft.

The power of the diode laser is monitored and recorded continuously/simultaneously by using a photodiode. Later, the NO_2 signal is generally normalized by using the photodiode signal. This is a regular approach for any LIF instrument and has been described previously by many studies. It is noteworthy that the impact of the correction was not significant during the field campaign PARADE. This is because during PARADE-2011, frequent calibrations were performed. So any variability in the power of the laser was captured via the calibration.

“**Line 157-265:** The calibration system that uses the NO titration by O3 to produce NO2 is described and used in different ground-based instruments (i.e. Ryarson et al, 2000, Matsumoto et al., 2000, Osthoff et al., 2006). In my opinion it is a good approach that can be a system for periodical laboratory check of the instrument performance and of the possible NO2 cylinder degradation, but according also to figure 7 it is the bigger part of the system and includes many components not so compact such us the ozone generator and the ozone analyser. The use of this calibration system seems not easy on ground-based field campaign and really complicated on aircraft.”

Response:

The formation of NO_2 via the gas phase titration is very common approach used for the calibrations of NO_x analysers. A reference to the previous study (Ryerson et al., 2000) is included. We agree with the reviewer that the bigger part of the instrument is the calibration system. However, most parts of the system (like MFCs, valves, reaction chamber, etc.) including the ozone generator is part of a single 19-inch rack mount (4RU). The ozone analyser is also 19-inch rack mount (4RU). So basically, two 19-inch rack mount and a small pump are required for the complete system.

The calibration system can be used to check degradation/changes in the concentration over a period of time in a NO_2 cylinder. We adapted such an approach in the past, but the day to day variation in different NO_2 cylinders was hard to track, since these cylinders showed unstable concentrations with low repeatability even within a short period of time. These checks were performed with different instruments (CLD, CRD, and GANDALF) during different periods of time. In a short time scale (hours), the observed difference was within 3-13% for different NO_2 gas cylinders. Whereas for a longer period (months), the differences were roughly up to 30%. Therefore, to get a reliable signal, the gas phase titration is advantageous compared to the use of a NO_2 cylinder.

“Technical corrections:”

“**Line 52:** It is quite rare but sometimes NO2 can be more than 100 ppb so I would replace ‘100’ with ‘hundreds’. Line 73: Add ‘the’ between ‘in’ and ‘past’.”

Response:

Replacement is added.

“**Line 74:** remove the subscript to the ‘v’ of pptv.”

Response:

It is done.

"**Line 79:** The reference reported (Dari-Salisburgo et al, 2009) describes the first ground-based system developed by that group, I suggest to substitute this reference with the work of the same group (Di Carlo et al., 2013) that reports the evolution of their TD-LIF for aircraft measurements that has better sensitivity and performances."

Response:

Added the reference "(Di Carlo et al., 2013)" at this position.

"**Line 182:** Remove 'Figure 3' "

Response:

It is done.

"**Line 710 (Table 1):** I would include the evolution of the instrument described by Dari-Salisburgo et al, 2009, used also for aircraft measurements, because it uses another laser a Nd:YVO4 pulse laser, and has better performance in terms of LOD compared with that described in Dari-Salisburgo et al, 2009, more details can be found in Di Carlo et al., 2013."

Response:

The successor instrument is indeed better in sensitivity from the predecessor. The overview of the instrument described in (Di Carlo et al., 2013) is also added to Table 1 as suggested by the reviewer.

References

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